

REQUEST FOR RECONSIDERATION

Claims 1, 4, 6-8 and 10-16 remain active in this application.

The claimed invention is directed to a hologram recording material and a process for producing same.

Hologram recording materials are typically valued based on properties of a high refractive index change at the time of recording, a high sensitivity, low scattering, environmental resistance, durability, low dimensional changes and high multiplicity. Issues as to environmental resistance and durability has focused on inorganic matrix materials as well as hybrid materials. Issues with inorganic hybrid matrix materials have focused on the uniformity of film formation, compatibility with the optically active monomer, curing speed and refractive index. Research continues into hologram recording materials having good properties.

The claimed invention addresses this problem by providing a hologram recording material comprising a metal oxide matrix and a photopolymerizable compound having an aromatic ring, wherein the metal oxide matrix has a halogenated C₁₋₄ alkyl group and an organic group containing no halogen. Applicants have discovered that the presence of a halogenated C₁₋₄ alkyl group in the metal oxide matrix which also contains an organic group containing no halogen provides a material having a high refractive index, high sensitivity, low scattering, environmental resistance, durability, low dimensional change, high multiplicity and compatibility with a polymerizable monomer. Such a hologram recording material is nowhere disclosed or suggested in the cited references of record.

The rejections of claims 1, 4, 6-8, 10, 12-14 and 16 under 35 U.S.C. §103(a) over Chandross et al. EP 938,027 in view of Loy et al. *Chem Mater*, Shustack et al. U.S. '566 and Hiruma et al. U.S. '831, of claims 1, 4, 6-8, 10 and 12-16 under 35 U.S.C. §103(a) in further

view of Otaki et al. and of claims 1, 4, 6-8, 10-16 under 35 U.S.C. §103(a) in further view of Poutasse et al. '782 or Zeldin et al. U.S. '172 are respectfully traversed.

None of the cited references of record disclose or suggest the claimed hologram recording material having a halogenated C₁₋₄ alkyl group nor the improved compatibility between the matrix and the photopolymerizable monomer resulting there from.

Chandross et al. EP '027 has been cited for a disclosure of a holographic recording medium based on methyltriethoxysilane, phenyltriethoxysilane, lauryl acrylate and a photoinitiator which is hydrolyzed to form a holographic recording material (examples 1-3). EP '027 corresponds with the disclosure in JP A-11-344917 discussed on page 3, lines 1-18 of applicants' specification and is reproduced below:

[0005] JP-A-11-344917 discloses an optical recording medium wherein an organic-inorganic hybrid matrix contains an optically active monomer. In the organic-inorganic hybrid matrix, a metal element has an alkyl group (a methyl group) or an aryl group (a phenyl group). However, the introduction of the methyl group makes it impossible to improve the compatibility between the hybrid matrix and the optically active monomer. The introduction of the phenyl group gives a more improvement in the compatibility than the introduction of the methyl group. However, the introduction of the phenyl group causes a fall in the curing speed

of a hybrid matrix precursor ([0015] in the above publication), and causes a rise in the refractive index of the hybrid matrix. If the refractive index of the hybrid matrix becomes high to approach the refractive index of the optically active monomer or a polymer therefrom, a high refractive index change is not easily obtained at the time of recording. This fact makes the flexibility in recording medium design narrow.

Above [0015] corresponds to [0016] of EP 938 027.

While the reference discloses an alkyl (a methyl group) or aryl group (a phenyl group) and that **the organic components** provide compatibility with the organic components (the

photoactive monomer) of the photoimageable system, there is no disclosure of a halogenated C₁₋₄ alkyl group. Thus, Chandross alone fails to suggest the claimed invention as the disclosure fails to suggest halogen substituted C₁₋₄ alkyl groups.

Loy et al. has been cited for a disclosure of sol-gels with halogen containing organic groups and that chloromethyl and chloromethylphenyl groups **react more slowly** than methyl, the **pore diameter** of chloromethyl is slightly **larger** than for that of methyl and that hydrogen, methyl and chloromethyl are **more transparent** than ethyl, dodecyl, hexadecyl, octadecyl and vinyl (page 3629, Table 6).

The official action asserts that it would have been obvious to one of ordinary skill in the art, based on the disclosed transparency in Loy et al. to have replaced the methytriethoxysilane of EP '027 with a chloromethyltriethoxysilane as disclosed in Loy et al..

In spite of Loy et al. measuring the transparency **of the gel** itself, and not for a photorecording medium containing a photoactive organic monomer, the assessment in the office action as to the effects of halogen substitution have been somewhat distorted, as the effects of “reacts more slowly”, larger pore diameter” and “transparency” are gross generalizations for which there is evidence to the contrary.

As to the reduced rate of reaction resulting from addition of halogen substitution, the examiner is respectfully requested to view the entirety of the data in Table 5, reproduced below:

Table 5. Gel Times for Organotrialkoxysilanes (Where Possible, Gel Times Are Given for 1.0 M Monomer Concentration; in All Experiments, Monomers Were Reacted with 1.5 equiv of Water)

| R group | trialkoxysilyl group, Si(OR') ₃ | | | |
|------------------------------|--|-------------------------------|---|------------------------------|
| | R' = Me | | R' = Et | |
| | conditions | gel time (±5%) | conditions | gel time (±5%) |
| OR' | H ⁺ , 1.0 M ^b OH ⁻ , 1.0 M H ₂ O, 1.0 M ^c | 60 days 12 h 64 days | H ⁺ , 1.0 M H ₂ O, 1.0 M ^c | 300 days 148 days |
| H | H ⁺ , 1.0 M ^d H ₂ O, 1.0 M ^d | 15 h 15 h | H ⁺ , 1.0 M H ₂ O, 1.0 M ^d | 2 days 2 days |
| methyl | H ⁺ ^a , 5.9 M OH ⁻ , 1.0 M ^d | 12 h 12 h | H ⁺ , 4.4 M OH ⁻ , 1.0 M ^c | 9 days 12 h |
| ethyl | | | OH ⁻ , 2.0 M ^c | 12 h |
| vinyl | H ⁺ ^a , 5.6 M OH ⁻ , 1.0 M ^c | 12 days 4 days | H ⁺ , 4.2 M OH ⁻ , 1.0 M ^c | 8 days 7 days |
| <i>n</i> -hexadecyl | H ⁺ , 1.0 M ^e OH ⁻ , 1.0 M H ₂ O, 2.4 M | 12 h 12 h 12 h | H ⁺ , 1.0 M ^c OH ⁻ , 1.0 M | 10 min 5 min |
| <i>n</i> -octadecyl | H ⁺ , 1.0 M OH ⁻ , 1.0 M ^c H ₂ O, 1.0 M ^f | <1 min 4 min 17 days | H ⁺ , 1.0 M ^f OH ⁻ , 1.0 M ^f H ₂ O, 1.0 M ^f | <<1 min 2 days <<1 min |
| chloromethyl | H ⁺ , 4.35 M OH ⁻ , 1.0 M ^b H ₂ O, 1.0 M | 77 days 26 days 87 days | H ⁺ , 1.0 M ^g OH ⁻ , 1.0 M ^c H ₂ O, 1.0 M | 172 days 2 h 76 days |
| <i>p</i> -chloromethylphenyl | H ⁺ , 4.0 M OH ⁻ , 1.0 M ^c H ₂ O, 4.0 M | 36 days <1 min 15 days | | |

^a 0.1 M HCl used. At neat concentrations (not shown), these monomers produced the results found in *b-g*. ^b Very exothermic, solution sprayed out of vial. ^c Nonhomogeneous gel composed of a mixture of precipitates and gel. ^d Exothermic reaction upon addition of aqueous catalyst caused bubbles throughout gel. ^e Phase separation of heavy oil or gum from solution. ^f Formation of precipitate. ^g Phase separation and formation of hard resin.

Thus, while the office action has accurately characterized the rate of gel formation to be slower for the halogen substituted organotrialkoxysilane, it is noted that the gel times for the chloromethyl species ranged from **26 days to 87 days** for the methoxy compounds and **2 hours to 172 days** for the ethoxy compounds, depending on the hydrolysis conditions. It is further noted for the corresponding methyl organotrialkoxysilane, the gel times was observed as **only 12 hours** for the methoxy compound and **12 h to 9 days** for the ethoxy compounds.

The acidic hydrolysis was increased from 154 to 344 times, between methyl and chloromethyl. Alkali hydrolysis conditions either reduced gel time or increased gel time 54 times. When the gel formation time is measured in days, such an increase can not be viewed as providing any motivation to use.

Moreover, under neutral hydrolysis conditions the chloromethyl compound demonstrated a gel time of from **76 to 87 days**. It is further noted that Chandross merely describes “mild cure conditions” (column 6, line 36) and only exemplifies in example 1 conditions of a solution of partially hydrolyzed precursor in acetone. Thus, the significance of the relative reduction in reaction rate has been “muted” in the official action such that use of such substitution may not have been considered to be obvious in terms of the negative kinetic effect in gel formation time.

Furthermore, as to the effect on transparency, while the reference and the office action assert that H, Me and chloromethyl were **the most transparent**, the transparency is apparently dependent on the hydrolysis method. Table 6 is reproduced below:

Table 6. Appearance of Monolithic Polysilsesquioxane Gels (Entries with Dashed Lines Indicate No Gels Were Obtained)

| R | RSi(OMe) ₃ | | | RSi(OEt) ₃ | | |
|---------------------------------------|-----------------------|-----------------|------------------|-----------------------|-----------------|------------------|
| | H ⁺ | OH ⁻ | H ₂ O | H ⁺ | OH ⁻ | H ₂ O |
| H | transl. | transl. | transl. | transp. | transp. | --- |
| CH ₃ | opaque | transl. | --- | transl. | opaque | --- |
| CH ₃ CH ₂ | --- | --- | --- | --- | opaque | --- |
| dodecyl | --- | --- | --- | opaque | opaque | --- |
| hexadecyl | opaque | opaque | opaque | opaque | opaque | --- |
| octadecyl | opaque | opaque | opaque | opaque | opaque | opaque |
| vinyl | opaque | opaque | --- | transl. | opaque | --- |
| ClCH ₂ | transl. | opaque | transp. | --- | opaque | transp. |
| (<i>p</i> -ClCH ₂)phenyl | --- | opaque | opaque | --- | --- | --- |

Clearly, the formation of opaque gels under alkali hydrolysis conditions, would not be considered acceptable for a holographic system and such an opacity would give one of ordinary skill in the art reason to pause as to the inclusion of such a monomer in a gel matrix.

Even further while the examiner has noted a larger pore size for the chloromethyl compound, some gels were nonporous. Tables 7 and 8 are reproduced below:

Table 7. Surface Areas and Mean Pore Diameters for Different R Groups in RSi(OMe)₃-Based Polysilsesquioxane Xerogels

| catalyst | surface area (m ² /g)/(pore diameter (Å)) | | | | | |
|------------------|--|---------------------|------------------------|------------------------|------------------------|------------------------|
| | OMe | H | Me | vinyl | ClCH ₂ | ClCH ₂ Ph |
| H ⁺ | 246/22 ^a | 617/67 ^a | nonporous ^c | nonporous ^c | nonporous ^c | nonporous ^c |
| H ₂ O | 342/50 ^a | 393/45 ^a | no gels | no gels | 28/380 ^a | nonporous ^c |
| OH ⁻ | 212/65 ^a | 566/40 ^b | 566/45 ^a | 70/407 ^a | 20/750 ^a | nonporous ^a |

^a Gels were obtained at 1.0 M monomer concentration. ^b Monomer reacts violently, boiling the solution. ^c Gels were obtained at neat monomer concentration.

Table 8. Surface Areas and Mean Pore Diameters for Different R Groups in RSi(OEt)₃-Based Polysilsesquioxane Xerogels

| catalyst | surface area (m ² /g)/(pore diameter (Å)) | | | | |
|------------------|--|---------------------|----------------------|------------------------|------------------------|
| | OEt | H | Me | vinyl | ClCH ₂ |
| H ⁺ | no gels | 766/40 ^a | 226/56 ^c | nonporous ^c | nonporous ^c |
| H ₂ O | 637/84 ^a | 662/41 ^a | no gels | no gels | 51/240 ^a |
| OH ⁻ | no gels | 502/49 ^b | 281/214 ^a | 140/338 ^a | 63/478 ^a |

^a Gels were obtained at 1.0 M monomer concentration. ^b Monomer reacts violently, boiling the solution. ^c Gels were obtained at neat monomer concentration.

Certainly, the motivation as to an expectation of larger pore size is mitigated by the observation of nonporous systems.

In short, while the official action has accurately reported **some of the evidence** from Loy as to gel formation of a chloromethyl substitution, negative evidence as to the effect of chloromethyl substitution has been conveniently ignored. Fortunately, the standard for obviousness is “the prior art as a whole” such that the reliance on merely the favorable evidence is improper. Certainly, upon review of the totality of the effects of the chloromethyl group in terms of significantly extended gel times, the occurrence of opacity and the formation of nonporous gels would not make it reasonable to expect that the chloromethyl group would be beneficial.

According to the claimed invention, introduction of a halogenated C₁₋₄ alkyl group into a transparent metal oxide matrix makes it possible to obtain a hologram recording material having a remarkably **improved compatibility between the matrix and the**

photopolymerizable compound having an aromatic ring and a remarkably improved compatibility **between the matrix and an organic polymer** generated at the time of recording (see page 5, lines 3-9 of applicants' specification). Namely the resultant hologram recording material having an improved compatibility is transparent, and the transparency of the material is maintained at the time of recording. There is no suggestion that a halogenated C₁₋₄ alkyl group would be more transparent than non-halogenated group or any suggestion of an enhanced compatibility between the gel and the photopolymerizable monomer or a polymer generated by polymerization of the monomer.

Page 4 of the official action has taken the assertion of an enhanced transparency of the matrix *per se* and concluded that such enhanced transparency would be expected "to confer at least some increased transparency to the holographic article."

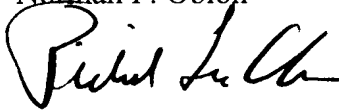
The examiner is reminded that the transparency of a holographic article is based in-part on the coexistence of the matrix and the monomer and that even if the matrix were completely transparent, an impairment in the compatibility with the monomer would not allow for the formation of a transparent hologram material. Thus, the examiner's assertion of an expectation of an increase in transparency based on the properties of the matrix alone, are simply not reasonable.

As the cited combination of references neither discloses or suggest the claimed combination nor the improvements in a holographic recording material resulting there from, the claimed invention is not rendered obvious from the cited references and withdrawal of the rejection under 35 U.S.C. §103(a) is respectfully requested.

Applicants submit that this application is now in condition for allowance and early notification of such action is earnestly solicited.

Respectfully submitted,

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